WORLDWIDE STATUS OF NUCLEAR APPLICATIONS TO THE WOOD, PAPER AND PULP INDUSTRIES.*

by

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Scope

The purpose of this paper is to review the applications and uses of isotope and radiation technology in the major foreign countries of the world in their wood, paper and pulp industries, and to thereby help define directions of profitable use of these new techniques for their American counterparts. This paper will not cover U.S. developments in these areas in detail, since these topics are the subject of all the remaining papers of this conference. The information contained herein is compiled from some 70 foreign references in this field, obtained through the cooperation of the Library of Congress and the U.S. Department of Agriculture. Much of this information is available from the Library of Congress which monitors the world foreign literature in radioisotopes and radiation technology on a continual basis, under contract to our Division. The product is in the form of a 3" x 5" file card -- one article per card, abstracted and translated. Some 1,000 articles are thus available per year at an annual subscription rate of $50 per year.[1]

Introduction

Forests covering about 30% of the world's area are ecosystems which constitute very complex interrelations between native plants and animals,

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litter and soil, and whose chief importance for man lies in the sum total of timber resources and forest products.

The importance of the wood, paper and pulp industries in our national economy is given by the following facts: 30 billion dollars of the 600 billion dollars of the estimated U.S. Gross National Product relate to the timber-based economic activities.[2] The value of the stumpage (out-standing timber) is estimated at one billion dollars to which two billion are added in harvesting. The production value of primary timber products (lumber, veneer, plywood, railroad ties, floorings, etc.,) is estimated at five billion dollars and that of secondary timber products (pulp, paper, paper-board, millwork, cooperage, containers, prefabricated wood products, furniture, naval stores, charcoal, wood distillation products and chemicals, cellulose plastics and fibers, lignin, etc.) at seven billion dollars.

Construction work of wood is estimated at eight billion and marketing and transportation costs at seven billion dollars. The national importance of timber-based economic activities can also be judged from the fact that of 69 million employed people more than three million were engaged in 1963 in timber-based activities.[3]

However large these figures appear, the impact of forest resources on the economies of countries such as Canada, Sweden, Finland, the USSR, and Brazil is much greater so that a rational utilization of forest resources, including the use of nuclear radiation and radioisotopes, should play a significant role in these countries and, in addition, could contribute a great deal to uplifting the economies of poverty-stricken tropical countries possessing rich forest resources.

In sharp contrast to the value extracted from our forests stands the waste involved in their utilization since only 60% of the cut stumpage is converted into useful products and the rest lost as waste or is unmarketable. Of the total forest lands, in the United States, of 760 million acres only 50 million are commercial forests and of the latter 20% (mostly hardwood forests) have so low grade timber that -- again, under the prevailing condition -- it does not pay to cut and market it. Thus, approximately 47% of our total timber resources must be considered unproductive at the present time. Although some of this land may have some recreational value, most of it occupies space that could be used for growing useful forests or for other agricultural utilization.[4]

The problem of the useless forest stands is given a major consideration in this paper and it may be hoped that it could be at least partially solved by new uses found for unmarketable timber through the application of high energy radiation. As a matter of fact, use of radioisotopes and radiation has the potential of increasing the yield and improving the quality and usefulness of practically all forest products since the effects of radiation on these lignocellulosic materials is rather pronounced.[5]

Radiation can be used at all stages of growth, starting from the seedling through forest growth, harvest, and the manufacture of primary and secondary forest products until the finished paper or textile fibre to which it could impart special desirable characteristics. Actually the application of radioisotopes or radiation should and does begin even before the forest is planted. Soil humidity can now be checked by numerous methods using neutron absorption. The presence and amount of essential nutrients

present only in trace amounts can be established by neutron activation analysis. The physical properties of forest soils, such as compaction or density, can be explored by means of gamma radiation.

The very important wind pollination of coniferous trees can be studied by tagging the pollens with phosphorus-32 via biosynthesis and setting up pollen traps, the activity of which would indicate their average dispersion.[6] The exchange of subterranean materials and translocation of nutrients have already been studied by Swedish scientists[7,8], while American scientists have investigated the breakdown of forest litter with radioisotopes.[9]

While almost all segments of the timber based industries have used radiation and radioisotopes, the widest use of these new techniques is made by the pulp and paper industry of this and many other industrially progressive countries. Among the 20 major industries of the United States using radioisotopes the paper industry is one of the leaders in the application of nuclear radiation and radioisotopes for industrial purposes, according to the 1959 study made by the National Industrial Board in New York and as reported in a Canadian Journal.[10] In a 12 month period, during 1957-58, the use of radioisotopes was reported by 99 paper manufacturing companies of which 93 achieved considerable saving (almost $3,000,000) through the use of radioisotopes. While the market value of paper and allied products has increased only about 25% above the high level of 10 billion dollars from 1957 to the present time, the application of radioisotopes has, according to estimates of paper specialists in the U.S. Department of Commerce, at least doubled and attained a two-fold increase in the savings.[11]

For this reason the present paper, while covering the use of radiation and radioisotopes in the most important timber-based industries, will place major emphasis on developments in the pulp and paper industry. This industry has been a pioneer in the industrial application of nuclear science.

In this paper the following subject areas will be specifically covered: Application of Radioisotopes and Radiation in Silviculture and Lumbering, Gamma-Ray Irradiation of Wood, Radiological Examination of Wood Products, and the Use of Radioisotopes and Radiation in the Pulp and Paper Industry.

APPLICATION OF RADIOISOTOPES AND RADIATION IN SILVICULTURE AND LUMBERING

1. Determination of Age of Trees

The peculiar feature of silviculture is the fact that its main object, namely the stand of living trees, constitutes both productive machinery (capital) and produced yield (interest). Careful judgment must therefore be exercised to obtain the optimum yield without diminishing the capital. This goal can only be attained through maintenance of a growing stock with correct age, species, diameter, and quality distribution of the trees. Only in this way will the best yield be obtained in perpetuity for harvesting. In order to provide a stand maintained for an 80 year rotation period, the age of the trees must be known. An exact measurement of the elapse of time can be attained with use of radioisotopes; this method was introduced by Willard C. Libby who determined the age of natural and man-made products by determining their carbon-14 content.[12] Although the decay period of radiocarbon is much too long (5760 years) and applicable only to organic materials that no longer assimilate, shorter lived radioisotopes such as tritium - with a more

convenient half-life of 12.36 years[13] could be injected into trees to help determine age of trees non-destructively.

2. Determination of Wood Soundness, Grading and Harvesting

The health, vigor, and quality of standing timber cannot always be detected by its outward appearance since the living tree may not show any defects and yet could be rotten in the core. To cut a healthy tree that should not be harvested only to see whether it may be diseased would defeat its own purpose. Consequently only non-destructive testing should be used to discover invisible defects. In this country, X-rays were used first for this purpose.[14] Radiographical techniques, developed later, allowed not only the detection of cavities and structural alterations caused by disease, fungi or insects but also a careful analysis of the opacity of the wood tissues to gamma-rays gave an insight to understanding the normal and pathological anatomy of the tree and permitted further determination of the structural quality of the wood and its taxonomic and industrial characteristics.[15] In spite of its usefulness the X-ray method has many shortcomings, such as: the distortion of the picture due to the pattern of the X-ray field and the cylindrical shape of the tree plus the need for electrical current supply and related heavy power supply which cannot be easily transported within very dense forests. The subsequent development in 1956 of a method for nondestructive testing of living trees by gamma radiography by the Japanese scientist Noriyoshi Lizuka removed the greatest impediments to the use of tree irradiation techniques in forest research and management.[16] This study was made to determine whether trunks of Todo-firs (Abies mayriana) are decayed in their natural state. Using gamma-rays of Co^{60} and a portable ratemeter he irradiated tree trunks at different heights and under different

angles and used the formula:

$$\bar{D} = \frac{n \cdot A - \sum_{1}^{n} B_n}{\sum_{1}^{n} C_n}$$

where $\bar{D}$ is the mean count number, n the number of observations, A the count number through air, B_n the n-th count number through wood, and C_n the penetration distance of the gamma-rays in the wood. In this way he determined not only whether the tree trunk was healthy or rotten, but also the extent and location of the rotten area as well as cavities conspicuous through rapid decline in density. Subsequent visual examination of 34 gamma radiographed trees revealed the high reliability of the method.

Two years after Lizuka's discovery Wayne B. Parrish established, in a Ph.D. thesis submitted in 1958 to Ohio State University,[17] the greater usefulness of such gamma-emitters as tin-113, iodine-129, ytterbium-169, cesium-137, cerium-144, and samarium-144, for non-destructive testing of living trees by radiographic means. Experimenting on oak trees with Geiger counters, scintillators, and radio-photography with emitters in the energy range from 23 to 561 KeV he found that emitters with energies between 50 and 150 KeV give the clearest picture of eventual irregularities within the trees and that scintillation counters with NaI(TI) crystals were best suited for detection purposes.

Although originally conceived as a means of non-destructive testing of living trees, the principle and methods of gamma-irradiation were soon extended for the inspection of used timber such as the structural wooden

parts of bridges or houses, trestles, sleepers, poles, beams, mine and tunnel shorings, etc., the soundness of which could be determined in place without boring. Portable timber analysers ("Densitometers") containing 200 microcuries of cesium-137 as a radiation source and a mercury cell powered electronic unit, weighing about 12 lbs., are now manufactured in this country and are commercially available for the determination of the soundness, density, and thickness of wooden materials.

The application of gamma-radiation to living trees will no doubt usher in a new era not only in silviculture but in plant physiology and plant taxonomy since with the help of this method the growth of trees can be observed in vivo in minute detail, differences in grain structure can be detected, and pathological changes, insect and fungus damage can be discovered long before visible symptoms are apparent and irreparable damage is done. Further, the forester now has a readily usable tool with which to determine which timber is ready for harvesting or culling.

3. Control of Cutting Efficiency of Machines

In modern forest management trees marked for cutting are sawn by machines containing serrated cutting blades such as chainsaws, bush combines, high-above-harvesters which are subject to considerable wear and tear. After mechanical or hydraulic debarking the logs are reduced to proper size in sawmills by band or circular saws, while veneer is made by rotary peeling or slicing. Reduction of wear and tear and maintenance of cutting efficiency may greatly reduce harvesting costs. For the control of wear and tear of cutting machinery numerous nuclear techniques are used such as the activation of

cutting blades by neutron bombardment or the insertion of machine parts containing trace amounts of the radioisotopes cobalt-60, nickel-63, manganese-54, etc.[18,19] By these means, reduction in efficient cutting or in operation is more easily detected and optimum speeds can be more easily controlled.

4. Smoke Detection

Forests, sawmills, lumber yards contain highly inflammable material which, especially in dry weather, constitutes a great fire hazard. We all deplore the tremendous losses of our forest resources caused by forest fires due not only to human negligence and carelessness but also due to natural causes. The prevention -- or at least a great reduction-- of these losses could be achieved, if an incipient fire could be immediately detected and extinguished before it reaches dimensions that are difficult to control. Radioisotopes may bring the solution of this problem and thus be a great blessing to our national economy and conservation of our natural resources. The phenomenon of ionization allows in a very simple way, the sounding of an alarm in time or the drawing of attention by other means to the imminent danger. Before fire starts in wooden material smoke evolves which can be drawn into a device and interferes with the ionization current.[20] For this purpose two small metallic ionization chambers (each one containing a small amount of radium or polonium) are in electrical counter-connection and are shunted by a triode to a cold cathode which is fed by a current of 220 V potential which operates the alarm relays. One chamber is closed, the other open, but under normal conditions the two chambers are in equilibrium. As soon as precombustion gases enter into the open chamber, the gas molecules interpose themselves between the ions which thus have time to recombine before

the collecting electrodes. This results in a rapid dimunition of the ionization current and by electronic relays sets the alarm mechanism in operation. Such devices are now available in this country and operate reliably from a 110 V., A.C. supply. A string of these can be set up in remote or sensitive areas to work independently or to augment visual procedures, particularly for night detection of smoke.

GAMMA-RAY IRRADIATION OF WOOD

1. Conversion of Wood into Edible Material

While the gamma-irradiation of wood for the purpose of inspection and testing requires only minimal radiation doses (of the order of 10^{-3} to 10^{-6} roentgen), the alteration of the physical and chemical properties of wood materials requires radiation doses many billions of time larger (of the order of 10^{8} roentgens) to produce chemical changes in the wood structure that are of practical interest. In order to understand the relatively high resistance of wood to both chemical and radiochemical attack we must consider the composition of this important and ubiquitous organic product. Its composition varies over a wide range, but is essentially 50 to 60% cellulose, 25 to 30% lignin and the rest is hemi-cellulose, water-soluble carbohydrates, gums and resins. In a simplified way we may say that wood consists of cellulose bricks held together by lignin mortar. While cellulose is easily hydrolysed and radiolysed, lignin is extremely refractory and, as a matter of fact, forms a shield that protects the cellulose units from radiolytic destruction by radiation intensities that could destroy other organic matter.

While the chemical composition of cellulose was readily discovered by way of acid or enzymatic hydrolysis to be a macromolecule consisting of an ordered chain of 6000 to 9000 glucose units, the constitution of lignin

resisted all pure chemical exploration and yielded only to methods employing radioactive tracers, especially carbon-14. It was the old master of wood chemistry and Director of the Institute of Wood Chemistry at the University of Heidelberg, Professor Karl Freudenberg, who at the end of a lifes work of wood exploration finally resorted to radiochemical methods to untangle the highly complex structure of lignin. He injected synthetic compounds that were suspected to be the precursors of lignin into the living plant and determined by step-wise degradation and radiochemical analysis of the obtained breakdown products whether the injected product was used in lignin synthesis.[21] When the injected compounds were tagged at definite places with carbon-14, the location of the carbon-14 atoms in the breakdown products allowed the reconstruction of the path of biogenesis of lignin and elucidation of its structure. This was established to consist of phenylpropane derivatives, especially coniferylalcohol[22] and shikimic acid.[23,24] This explains the radiation resistance of wood since it is well known that aromatic structures of this kind yield only very reluctantly to radiation, but once they have become radiolysed tend to break down completely. Similar investigations were performed by the Austrian scientists G. Billek and K. Kratzl who studied the lignin of spruce (Picea excelsa) with injections of hydroxyphenylpyruvic acid-3-C,^{14} thus carrying the biosynthesis one step further.[25]

Independently of these exploratory and analytical investigations American and Russian scientists tried to convert wood and wood waste by intense radiolysis into useful products. In this country Saeman and Millett of the U.S. Forest Products Laboratory, Madison, Wisconsin, in cooperation with

E. J. Lawton of the General Electric Research Laboratory, Schenectady, New York, bombarded wood and sulfite pulp with high energy cathode rays at a dose of 10^8 roentgens and achieved depolymerization and decomposition of the irradiated material.[26] In the USSR, A. S. Freydin of the Central Research Institute for Wood Working, Leningrad, and U. M. Malinski and D. Y. Karpow of the Physicochemical Institute, Moscow, explored the effect of ionizing radiation on wood.[27] They found: (a) that gamma-irradiated wood lends itself better to acid hydrolysis for glucose production as it requires only a diluted acid and reacts more readily, and (b) that irradiated wood is equally well assimilated by rumen bacteria as the usual cattle fodder and thus can be used to replace it to a certain extent in the nutrition of ruminants, provided small amounts of non-carbohydratic essential nutrients are added.

The Russian workers reduced wood from Pinus sylvestris into small pieces of 0.5 to 1.0 mm size and exposed them at normal temperature in both air and in vacuo to gamma-radiation from 600, 1400, and 20,000 curies of cobalt-60 at a dose rate of 500 x 10^6 roentgens per hour for 1 to 3 hours. After irradiation the wood was extracted with hot water and yielded some monosaccharides as shown in the following table.

Irradiation time, in hours	Content in water-extractable monosaccharides of wood after gamma-irradiation at dose rates 500 X 10^6 r/hr, in %				
	Galactose	Glucose	Mannose	Arabinose	Xylose
0	0.02	trace	trace	0.02	0.04
1	0.03	0.08	0.20	0.02	0.04
2	0.03	0.065	0.20	0.04	0.05
3	0.03	0.22	0.50	0.10	0.20

Subsequent hydrolysis with 1% hydrochloric acid rendered all cellulosic and hemi-cellulosic material water-soluble in form of monosaccharides.

In 1955 and preceeding the above Russian work, one of us (F. J. Weiss) predicted in an article, "Food for Tomorrow," that irradiation of the enormous amounts of lignocellulosic farm and forest wastes (straw, corncobs, bagasse, sawdust, etc.) accumulating every year could be converted into cattle fodder by way of irradiation.[28] He pointed out that the conversion of only 10% of this material into fodder for ruminants would add about 4.25 million tons of carbohydrates to the annual feed supply in the United States. Of course, to make this waste material fully utilizable in form of meat and dairy products about 2% of non-proteinous nitrogen, preferable in form of urea, could be added. However, not much research work has been carried out on the digestibility of irradiated herbaceous matter to increase the availability of animal protein, however important it is for an exploding world population whose diet is particularly deficient in nutrients of animal origin.[29]

2. Wood-Plastic Materials

In the field of plastics, the gamma-irradiation of wood looks very promising. Since ionizing radiation destroys the oxygen and C-C linkages that hold the cellulose and lignin moiety of wood so strongly together, the breakage of these linkages may produce on both the carbohydratic and the aromatic ends free radicals which, however, may rapidly undergo recombination reactions unless substances are present that are even more reactive. In fact, the presence of such substances in form of various monomers (vinylchloride, acrylonitrile, styrene, methylmethacrylate, etc.) at the moment of irradiation-destruction

may lead to instant graft polymerization and the formation of hard, durable, and fairly indestructible products that retain the outer appearance of the wood species from which they were made, but are quite different chemically and in their physical properties to make them suitable for numerous uses for which wood, as such, would not be suited. While there are many methods of grafting all kinds of monomers onto pure cellulose,[30] the application of irradiation or pre-irradiation grafting to wood offers most interesting and challenging possibilities. American scientists are, at the present time, actively engaged in the realization of this very promising new avenue of wood utilization,[31] while the Russian scientists seem also to be greatly interested in wood copolymerization by gamma-irradiation.[32]

In this connection, it should be mentioned that the lower irradiation doses that are needed for the scission of the ligno-cellulose complex in wood are sufficient to produce chemical changes in its structure (most likely inter-and intramolecular cross-linkage) which impart desirable physical and chemical properties to the wood. The Dow Chemical Company work in 1959 showed that radiation doses from 10^4 to 10^6 reps induced a significant hygroscopicity in small wooden blocks. While no difference was found in radial swelling tests, the tangential swelling of irradiated wood was markedly reduced compared to non-irradiated control specimen. Agar-block tests were made with the fungus Lenzites trabea which showed that irradiation below 10^6 reps reduced the decay susceptibility, while doses above 10^6 reps brought a remarkable increase of susceptibility -- as was to be expected.[33]

Bersenev and Fokina irradiated building boards made from sawdust with a resin binder at a rate of 2,300 to 35,000 roentgens per hour and showed that the stability of the boards, based on their water absorption capacity, was increased as the water absorbing power was reduced.[34] Hachihama and

advantage that the wooden material which may be the part of a house or a bridge can be tested in situ while the object of the test remains untouched. Of special interest is the moisture content of both fresh and dried wood and a method by which the drying process can be continuously recorded. Westermark, Sjostrand, and Forsberg developed a moisture measuring method for moving material in which two beta-ray transmission channels are electrically coupled against each other and properly balanced. The drying can be measured from the area-mass difference proportionate to the disappearance of water.[36] A similar process is described by Noack and Kleuters who used 20 millicuries of strontium-90/yttrium-90 and a scintillation counter, photomultiplier, and rate meter for the continuous measurement of the moisture content of veneer wood. This method can also be used for the automatic control of veneer drying and particle board manufacture.[37]

In contrast to the above methods using beta-emitters, the Finnish investigators, Kajanne and Hollming determined the moisture content in wood chips by means of gamma-ray scattering. Using a radiation source of 120 millicuries of cobalt-60 enclosed in a lead ball with a very small opening, they aimed the gamma radiation towards the axis of a cyclindrical target container of aviation plywood and measured the scatter with a scintillation counter and scaler. Since the Compton electron density is a direct linear function of the moisture content, the latter could be determined with an accuracy of 1%.[38]

2. Decay and Insect Damage Determination

Of special importance are nondestructive tests for the determination of decay resistance and insect damage of wood used in poles or in underwater constructions. K. Nietzsche of the Electrotechnical Institute in Ilmenau (East Germany) has developed a method for the determination of decay in

wooden poles which is based on the differential absorption for radioisotopically-tagged salt solution distributed between healthy and decaying wood. Since decaying wood absorbs much faster than healthy wood, a simple and reliable decay test could be made using iridium-192 salt solutions distributed between healthy and decaying wood. Since decaying wood absorbs much faster than healthy wood, a simple and reliable decay test could be made using iridium-192 salt solutions.[39] Marine piling which supports many docks and other marine construction is often infested with marine borers. Thus the piling becomes weakened and may give way under stress. It would be of great economic value to locate damaged piling in time and to replace it, if nondestructive methods could be developed to detect such damage. Some progress has been made in this direction. The Battelle Memorial Institute, Columbus, Ohio has developed a gamma-radiographic method using thulium-170 for the underwater inspection of 3.5 inches-thick waterlogged marine piling.[40]

In closing this section, two review articles should be mentioned which provide a good insight into the wide application made with radioisotopes and radiation, especially in the communist countries of Eastern Europe.[41,42] This includes the use of radio tracers in wood research and for the control of woodworking machinery.

THE USE OF RADIOISOTOPES AND RADIATION IN THE PULP AND PAPER INDUSTRY

The pulp and paper production is, with the exception of groundwood and other non-chemical processes, essentially a chemical industry and, therefore, many methods using radioisotopes as tracers or nuclear radiation for control and testing purposes are applicable in ways similar to their use in the chemical industry. This, for example, is the case for measuring the flow of

raw materials, determining reaction kinetics, analysis of intermediary and final products, etc. An annotated bibliography of all these methods, processes, and devices using radioisotopes is currently collected and pertinent articles from selected sources of foreign literature abstracted by the Science and Technology Division of the Library of Congress under agreement with the Atomic Energy Commission and can be found under the heading, "Chemical Industry," in the publication, "Radioisotopes in World Industry."[1] For this reason only those methods and devices will be discussed here which either are specific for the paper industry, such as measurement of area thickness and weight, or are of outstanding importance in paper manufacture.

The manufacture of paper can be conveniently divided into three stages: the pulping of wood which constitutes over 90% of the fibrous raw material; the bleaching of the pulp; and the interweaving of the fibers to produce the paper and allied products.[43] The raw paper may then be subject to quite elaborate finishing processes depending upon its ultimate use or may be combined with resins or plastics to form a large array of articles for innumerable uses.

The principal chemical pulping processes are the sulfite, the sulfate, and the soda process; the common purpose of each is to dissolve the lignin in order to reduce the wood to cellulose fibers.[44]

In the sulfite process the cooking liquor is made at the pulp mill by burning sulfur to sulfur dioxide and reacting the gas with limestone or milk of lime to obtain a cooking acid of desired strength. The wood chips are treated with this liquor in the digester at temperatures from 110° to 150°C from 8 to 12 hours. The main objective of this procedure is to obtain complete penetration of the chips by the liquor since incompletely penetrated

chips give low yield and require more bleaching. To observe this all-important phenomenon of penetration more closely, Arakin treated spruce and pine chips with sulfurous acid (containing calcium-45 and sulfur-35) at various temperatures, pressures, and times and obtained a very exact autoradiographic picture of the penetration process in the wood chips.[45] He demonstrated that steaming under pressure at 105°C greatly intensified the penetration of the chips. This was also dependent on the wood species and the time of harvest since the calcium-45 content of spring wood was higher than that of summer wood.

A less detailed but more general study of the sulfite process was carried out by the Japanese technologist, Ueno, who studied the thermodynamics and kinetics of the sulfite cooking process by means of cooking liquor tagged with calcium-45.[46] In a comprehensive study on "Radioisotopes in the Paper Industry," Cameron, of the Isotope Research Laboratory in Wantage (Berkshire, England) discusses the tagging of wood chips with very thin copper wire, labeled with copper-64. The powerful gamma-radiation emitted by this shortlived radioisotope allows one to follow the path of the tagged chips through the walls of the digester, to follow their slow disintegration, and to determine the removal of lignin.[47] Also Rutledge of the Mead Corporation, Chillicothe, Ohio, mentions among the various uses of radioactive tracers, the measurement and control of wood chip movement in digesters, break-up of ligno-cellulose and dissolution of lignin, flow of fiber suspension, pulp and water balance.[48] Radioactive calcium-45 was also used by Hostomsky and coworkers to detect the formation of carboxyl groups in cellulose during the pulping process.[49] They used tagged calcium acetate and measured the reaction

between calcium ions and carboxylic and sulfonic groups by the corresponding decrease of the activity of the solution containing calcium ions. They found that the latter attains a maximum at the moment of the start of solubilization of the lignin.

The most remarkable recent development in sulfite pulping was the replacement of the batch by the continuous pulping process. As long as calcium was used as base and an excess of sulfur dioxide was necessary to keep it in solution, this transition was not possible. It took the pulp and paper industry 70 years to learn that large amounts of SO_2 need not be recycled, when bisulfite in connection with soluble bases, such as magnesium or sodium, are used as cooking agents. This non-recycling procedure greatly simplifies the pulping operation, reduces the amount of capital required for SO_2 production, shortens the pulping time, produces a more uniform product -- and what is important from the viewpoint of radioisotope utilization -- allows a more rational application of tracers and radiation for process control.[50] Ljunggren describes in a comprehensive review article (devoted especially to recent developments in the pulp and paper industry of Canada, Sweden, and Finland) how wood chips can be tagged and then used for process control in continuous digesters.[51] He mentions three methods of flow-velocity measurements: (a) the passage method using two detectors located at a certain distance; (b) the total impulse method; (c) the attenuation method for measurement circulation of wood chips and cellulose in the digester during continuous operation.

No mention of radioisotopes or radiation was found in the literature in connection with the two other chemical pulping processes, namely the sulfate

(kraft) and the soda process. This may be because they have not yet reached the degree of technical refinement of the sulfite process or more probably, because some of the above mentioned tagging methods are applicable in analogous ways as in the two other pulping processes and this information is not published openly.

We encounter radioisotopes again in the pulp and paper manufacture at the pulp bleaching stage. The objective of bleaching is the production of brighter pulps as measured by light reflectance. Bleaching involves the removal of residual lignin and other coloring substances either by way of chlorination or by oxidation with calcium or sodium hypochlorite. In recent years chlorine dioxide has also been used for this purpose since it produces higher brightness without decrease in fiber strength.[52] Considering the multitude of bleaching agents and the different bleach requirement of various pulps, the "bleachability" test is of great importance for the determination of the exact amount of bleaching agent needed and the rational execution of the bleaching operation. The "Hypochlorite bleachability," for instance, is the amount of hypochlorite required for a given weight of pulp to obtain a standard brightness in a single-stage bleaching. Among other methods to determine the bleachability is a radioisotope tagging technique, using iridium-194 which is used for measuring the distribution of fibers in bleaching systems reduced either with copper or sodium formate.[53] In the latter case the pulp is soaked in a solution of tagged $(NH_4)_2IrCl_6$ and reduced with $HCOON_a$ for 14 hours at 90°C. The retention of activity during each bleaching state is measured and is found usually to be more than 90%.

The "paper stock" preparation comprises all intermediate operations between pulping and papermaking, that is, the preparation of the "furnish" -- a watery slurry of pulp fibers and chemicals -- and includes the beating and refinishing. In all these operations a microbiological control of the material is essential. This has been realized only recently when the troubles caused by microorganisms to the paper industry have become a matter of great concern to the paper chemists.[54] Micro-organisms (algae, bacteria, fungi) cause slime formation which clogs the pipelines and interfere with other operations, corrode the metal parts of the machinery, and even attack cellulose by breaking the chain molecules of the fibers. The control of these micro-organisms is, therefore, an important task of the pulp and paper technologist who constantly searches for better and more effective toxicants to get rid of this nuisance. For this reason experiments conducted in the laboratories of the Central Research Institute for Wood Structures in Moscow, USSR, were extended to include also the microbiology of wood pulp.[55] The effect of nuclear radiation on micro-flora and fauna-contaminated wood pulps was studied. The experiments were carried out with Coniophora cerebella which was tagged with phosphorus-32 or sulfur-35 in such a way that activities did not interfere with the physiological characteristics of the micro-organism in the pulp. This development, under normal condition and under the effect of various toxicants, could then be studied.

In the actual papermaking process batches of prepared stock are dumped into stock chests from where they are fed continuously at about 3 per cent consistency over finishing "jordans" (conical rotors revolving inside conical

shells at 300 to 550 rpm) into the paper machine system. After dilution to 0.5% and final screening the stock enters the wet end of either the Fourdrinier or the cylinder machine where the most essential part of the paper manufacturing process takes place -- namely, the sheet formation through agglomeration of the individual fibers. If the fibers are uniformly and randomly dispersed in the liquid, a well-formed sheet will be produced. If however, the fibers tend to flocculate and form clumps, the resulting sheet will be inferior. The most important thing for the papermaker is, therefore, to maintain optimum fiber dispersion throughout sheet formation and prevent tendencies to flocculation. Numerous techniques have been used to study flocculation and methods to prevent it, but it was not until the advent of radioisotopes and especially the extremely sensitive methods of micro-autoradiography that a clear scientific picture of fiber behavior was attained. Ljunggren explains the application of these methods[56] and he, as well as Casey[57] refer to the work of Allen and coworkers who developed methods of tagging fibers with iodine-131 in order to study flocculation and its effect on the final sheet.[58] From the headbox of the Fourdrinier machine the stock slurry flows through a "slice" onto a flat moving wire which is actually an endless belt and then over suction boxes to steam-heated dryer rolls, felt blankets and to final dryers and calendars. In this process more and more water is lost until the final sheet contains a well-controlled low water content. It is this last stage of paper formation that the thickness and area weight of the paper is determined and where nuclear radiation found its first use in the paper industry.[59]

While gamma-radiation is too penetrating to serve as area-weight or thickness gage in the paper industry, beta-rays -- and for certain purposes

even alpha-rays -- have a suitable penetrating energy. As far as beta-rays are concerned, there are two fundamental methods of area-weight determination in use: the first consists in the measurement of beta-ray absorption and requires the placement of radiation sources and radiation detectors on opposite sides of the paper; the second is based on beta-ray backscattering and allows the placement of both source and detector on the same side of the sheet for which the thickness or area weight is to be determined. Both methods give continuous measuring values as the paper strip passes by and lend themselves to automatic thickness or weight control.[60]

Although the energy loss of beta-rays in passing through matter is a complex physical process, the energy decrement can be expressed with an accuracy sufficient for practical purposes by the standard formula:

$$I = I_0 \cdot e^{-a.d}$$

where I_0 is the intensity of the incident radiation, I the intensity of radiation after its passage through the paper, a is the mass absorption coefficient in $(\text{meter})^2/g$ and d the area weightpenetrated in $g/(\text{meter})^2$.[61] The area weight in grams per square meter or in milligrams per square centimeter, has been used all along in the paper industry to express the paper strength. Since there are numerous radioisotopes available that could serve as beta-emitters, one would have tochoose one whose penetrating power stands in favorable relation to the area weight of the paper and this is the one whose half-value thickness lies in the same order of magnitude as the area weight of the paper. The radioisotope should also have a sufficiently long half life and specific activity (i.e. curies per gram) and should be available

in sealed ampoules.[62] Pankov and Kugushev examined the most readily available radioisotopes that meet the above specifications, namely H^3, C^{14}, S^{35}, Cl^{36}, Sr^{90}, Pm^{147}, and Tl^{204} from the viewpoint of their suitability for the measurement of paper strength and found that Tl^{204} and Sr^{90} gave the most satisfactory results in paper weighing.[63] The same was found independently by the Rumanian scientists, V. and S. Cristea.[64]

Although Geiger counters, scintillation counters, and ionization chambers can be used to measure radiation intensity, for the purpose of the paper industry the latter have generally been found most convenient. Several foreign firms that specialize in the manufacture of radiation equipment or in machinery for the pulp and paper industry (for instance, Frieseke & Hopfner in West Germany or Karlstads Mekaniska Werkstad in Sweden) supply complete paper weight measuring apparatus to be attached to the papermaking machines, especially to the Fourdrinier machine. These perform automatic weighing operations, which without the help of radioisotopes, would require much more time and labor and would yield less satisfactory results.

In most cases the paper manufacturer is not so much interested in the absolute area weight of the paper as it comes from the machine than in the relative value, or more correctly, in the deviation of the current weight from a certain standard value. For this purpose, niceties of geometry and atmospheric conditions can be dispensed with and the desired value fixed by a potentiometer fed by a constant current and connected with the ionization chamber using the compensation principle. An indicator shows the difference between the potentiometer and the ionization chamber voltage.[65]

For still greater precision in area weight determination Kuhn has proposed the measurement of the forward-scattering of Bremsstrahlung which increases proportionately to area thickness or density. On the basis of the metallic silver content of different exposed films, variable blackening intensities can be determined with great exactness which correspond to the measured irradiation intensities.[66] Also Nekhaevskii earlier described a further refinement of the automatically weight measuring apparatus using thallium-204 as a radiation source and a balancing circuit with a vibrating reed electrometer.[67] Bauche has explored the reasons for the uneveness of paper strength with the help of a Frieseke apparatus (FH 46) for area weight measurement and has stressed the economic advantages of continuous and automatic area weight measurement in the paper industry.[68] The Japanese scientist, Nagao, studied the influence of ambient temperature and air pressure on the accuracy of the reading by comparing the value of two movable weight-measurement apparatus with thallium-204 sources. These effects are not negligible, if high accuracy is desired. Thus, he installed a special air blower system and attained an accuracy of 1% in the sheet weight.[69] A similar device is described by another Japanese scientist who attached an air-blowing unit to the customary measuring device (beta-radiation source, ionization and reference chamber) which blows air through the path of the beta-rays. This device can weigh paper of 80 g/m^2 thickness and shows a drift of only 0.6 g/m^2 in 12 hours.[70]

Because of its continuous and automatic performance the apparatus described above, lends itself particularly well to the automatic control of paper quality and eveness. If the apparatus is attached, for instance, to

a Fourdrinier machine, any deviation from the set standard weight (if it exceeds a certain predetermined value) sets a relay mechanism in operation which opens or closes the stock valve, thus bringing about the necessary correction in order to maintain the desired area weight or paper thickness.[71] Such apparatus is available in the U.S. and will be discussed later in this conference.

The paper manufacturer is not only interested in area weight and thickness of paper but also in the uniformity of his final product. Uniformity is usually determined by visual inspection, that is, by looking at the light transmission through the final paper sheet. However, such subjective testing methods do not give quantitative data and, therefore, have been replaced by light source and photo-cell. But this latter method cannot differentiate between opacity caused by difference in mass distribution and the intrinsic opacity of the fiber which varies considerably. To overcome this problem, the Karlstads Mekaniska Werkstad in Stockholm, Sweden, devised an apparatus containing an ampoule with 65 microcuries of krypton-85, as a beta source. This is enclosed in a lead bar into which a hole has been drilled. The collimated beam of electrons passes through this hole onto the paper sample which is drawn past at a constant speed of 100 mm.per hour. On the opposite side of the paper is the end-window of a Geiger counter which is connected with a Philip's high-voltage supply and feeds pulses to a ratemeter. The 5 milliampere ratemeter output is then fed to a recording device.[72]

An interesting use is also made of alpha-rays to detect uneveness in paper. Because of their short but well-defined range these rays will penetrate thin paper only at locations where the thickness is less than the

alpha-ray range. A photographic film attached to the opposite side of the paper will reveal these areas by intense blackening and thus reveal variations over larger areas.[71]

Larger alpha-sources are also used in the paper industry to prevent the accumulation of static electricity which not only may lead to improper functioning of machinery but also involves an eminent fire hazard, especially when paper is treated with insulating or highly inflammable material. The alpha-radiation emitted ionizes the air and leads to the dissipation of the charge carriers through the air to the ground.[71]

The paper coming from the machine is not the final product of the paper industry. Sometimes elaborate processes are used to give it characteristics for a specific purpose. Paper may be filled or loaded with clay, titanium dioxide, calcium carbonate, talc, gypsum, zinc oxide, lithopone, barium sulfate, zinc sulfide, etc. The paper may also be sized with wax or sodium stearate, coated with starch, glue, resins, synthetic polymers, or laminated with plastic solutions. In all these operations radioisotope gaging may be applied to control the uniformity of application. A common process is the lamination of kraft paper with phenolic resins in the manufacture of formica. For good lamination the ratio of resin weight to total weight must be constant and precisely controlled. This can be done by using two beta-gages, one before the paper passes over a roller (the underside of which is immersed in a tank of liquid resin) and the other after it has left the curing oven. If both gages are synchronized, their connection in an electric circuit may indicate any deviation from the desired ratio.[71]

The direct grafting of monomers onto the cellulose fibers by gamma-radiation is a recent development in the field of paper industry.[73] This is

done to attach the coating more firmly to the paper and also to attain other effects. Cellulose has a very low resistance to high-intensity radiation, and becomes discolored and very weak. Attempts have been made to produce a more radiation resistant paper for certain purposes. Impregnation with acetophenone, naphthone, and phenol and subsequent irradiation have been to no avail. However, Glaudemans and coworkers discovered that benzylhydroxyethlycellulose films remained stable at irradiation intensities which destroyed pure or impregnated cellulose.[74] This apparently is due to the formation of a covalent bond between cellulose and the aromatic compound, a process similar to what is observed in nature, where the lignin serves as a shield to the cellulose in the irradiation of wood.

This chapter on paper should close on a sentimental note, an expression of gratitude by the chemist and especially by the nuclear chemist for those who have brought filter paper to the high degree of perfection as to keep step with the even greater refinement of research that nuclear chemistry has brought about. Mention was made of the various methods by which radiation and radioisotopes are increasing the efficiency and economy of paper production and the quality of its products, but fairness requires that also a word should be said about the contribution of the paper industry to the development of nuclear chemistry and radioisotopes. Nobody will argue that the lowly filter paper is still the backbone of analytical chemistry. In fact, the development of modern chemistry and the detection of radium would have been impossible without it. Names like Whatman or Schleicher & Schuell strike a familiar chord with every chemist. However, since the time when gravimetric analysis reached its highest perfection new and even more sophisticated and sensitive

methods were introduced to detect smaller and smaller amounts of substances such as trace elements or radioisotopes. Mention need be made only of paper chromatography, paper electrophoresis, auto-radiographic and activation analysis of chromatograms.[75] To meet the high specifications of these methods the manufacturers of filter and chromatographic paper reduced the ash content of their products. For an ordinary filter paper of 11 cm diameter the ash content is already as low as a few micrograms. This was reduced by several orders of magnitude[76] and this remarkable result was achieved without weakening the physical structure of the cellulose. These qualities are essential to obtaining a so-called high "R_f value" in chromatographic separation. The elimination of such ions as calcium, magnesium, ferric, chloride and sulfate allowed the identification of minute traces of elements on chromatograms by neutron activation analysis. Although a large literature on paper chromatography exists,[77] space forces us to restrict ourselves to the mention of three papers.[78,79,80] Although nothing can be found in the literature on the methods by which so high degree of purity could be achieved, there is little doubt that continuous control by neutron activation methods must have been applied.

Additionally, the quality of cellulose powder for column chromatography has been greatly improved. It is now widely used in the preparation of artificial radioisotopes. In this way the paper industry repays manifold the services it has received from radioisotopes.

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ADVANCES IN RADIOISOTOPE PRODUCTION AND UTILIZATION IN SCIENCE AND INDUSTRY

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INTRODUCTION

Only three decades after the discovery that radioactivity could be artifically induced, man-made radioisotopes have been established as powerful and versatile tools for an almost countless number of uses in nearly all fields of research and application. Man-made radioactivity is truly one of those discoveries of major benefit to mankind.

Advances in the discovery and production of specific radionuclides have been phenomenal. Although around 130 radioactive species have received significant usage and are routinely produced, the potential number of useful radioisotopes is much greater. Means of production cover a variety of accelerators with great flexibility in type and energy of particles and radiation generated, encompassing the use of photons, neutrons, protons, deuterons, tritons, helium-3 and alpha particles and heavier nuclei. However, the bulk production of radioactivity is now by means of nuclear reactors. Through neutron bombardment in high flux, high power reactors, production of most useful isotopes can be expanded to meet any conceivable need, up to hundreds of megacuries of certain isotopes such as cobalt-60. Also, vast quantities of fission products are unavoidably obtained as by-products of power and/or plutonium production. Individual radioisotopes from this source are being separated and purified in megacurie quantities. Costs of production have been greatly reduced and could become even lower with larger demand. These megacurie quantities of both neutron induced and fission product isotopes are being used in the development of heat source and radiation processing applications.

Advances in radiation detectors, associated electronics technology and the automatic recording and analysis of data also have been astonishing. Use of crystal scintillation counters has become routine and efficient crystals are available over 20 cm in diameter. Plastic detectors are also available in a wide variety of shapes and the sizes can be of meter proportions. Liquid scintillation counting has become extensively employed for efficient internal particle counting of a broad variety of sample forms. Use of liquid, plastic, and crystal detectors, with associated circuitry involving multi-channel, multi-parameter pulse height analysis, coincidence and/or anti-coincidence techniques, has been extended to extremely low level counting of samples, even to the size of human beings. Recently, a variety of semiconductor detectors have been developed that provide increased energy resolution for counting of alpha particles, conversion electrons and low energy X- and gamma rays. Semiconductor detectors incorporating heavier elements are under development to permit efficient, high resolution counting of higher energy gamma rays. Increasing attention is being given to automatic recording and to computer analysis and reduction of data from isotope and activation analysis procedures.

Parallel improvements and expansion in utilization of radioisotopes have taken place rapidly in all fields of application. In the basic physical sciences, the major applications are for analytical techniques, determination of molecular structure, measurement of reaction kinetics and determination of physical properties and behavior of matter. In the applied sciences, new uses are being developed in engineering and in such fields as oceanography, meteorology, hydrology, other geosciences and space technology.

In many instances, discoveries are being made through the use of radioisotopes which are so important and

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striking that we tend to lose sight of the fact that they would not have been demonstrable but for the availability of an abundant and varied supply of radioisotopes. The overthrow of the parity principle in weak interactions[1], the evolution of a generally accepted theory of Fermi interactions involving beta decay, muon decay, and muon capture[2,3], and the historic discovery of the Mössbauer effect[4] are all magnificent examples of what has been made possible by radioisotopes.

This paper will examine some of the major advances made in the above areas in the United States during the six years which have passed since the second United Nations Conference on the Peaceful Uses of Atomic Energy.

LARGE-SCALE PRODUCTION OF ISOTOPES

One phase that has surpassed all others in the United States in rapidity of growth since the last Geneva Conference is the development of processes for and the actual production of extremely large quantities of individual, purified fission products and neutron products. The incentive for this effort has come largely from the concurrent development of small electrical power supplies which utilize the heat of radioactive decay as their energy source. Another paper at this Conference[5] describes these power systems.

The fission product that has received the most attention for isotopic power use is strontium-90. In the period 1961-1964, almost five million curies of this isotope were recovered from waste streams and converted to the chemical form strontium titanate ($SrTiO_3$), a ceramic with good thermal and radiation stability, reasonably high strontium content, and relatively low dissolution rate in water[6]. This required the services of a large production site and two hot cell facilities.

Of the neutron products used for heat sources, plutonium-238 has been of greatest utility. It is produced in kilogram quantities by irradiation of neptunium-237, which in turn is produced by U^{238} (n,2n) and U^{235} (2n, gamma) reactions in reactor fuels. Three production sites and a large radiochemical laboratory are involved in the process. To date, all nuclear power supplies used in earth satellites have employed plutonium-238 as their energy source.

In our program to develop radiation for the processing of food and chemicals[7], we expect cesium-137 to find expanded use as a gamma radiation source. In anticipation of these applications, a 215,000 curie source of cesium-137 was completed in January, 1964, by Oak Ridge National Laboratory. The final source, doubly encapsulated in stainless steel, will be used in the radiation development program at Brookhaven National Laboratory[8]. Approximately one million curies of cesium-137 have been purified by a very simple process involving elution of a cesium fraction from an inorganic ion-exchange medium and subsequent purification by the alum crystallization process[9].

New opportunities for the use of cobalt-60 will arise from a program now under way to manufacture this radioisotope in specific activities of 400-500 curies/gram or even greater. A capability for the manufacture of hundreds of megacuries per year of cobalt-60 exists in the United States, although total current utilization is only about three megacuries. Tritium and carbon-14 are also available in quantities challenging the imagination of those of us who were early workers in the field. Such large capacities for the production of isotopes by neutron absorption arise from the increased availability of ideally suited reactor space resulting from curtailment of plutonium production in the United States.

Curium isotopes are receiving a great amount of attention at this time, for two reasons: they are part of the irradiation chains leading to the formation of the very heavy elements, and they appear useful as heat sources in various applications. Plans are being carried to completion to provide a facility for the processing of kilogram quantities of curium-244 into targets for further irradiation in neutron fluxes in excess of 10^{15} neutrons/cm^2 sec[10]. A special reactor is being built for the latter purpose, and it is scheduled for completion in 1965[11]. The product of the high-flux irradiation will be gram quantities of californium-252, for study of this element itself as well as for further irradiation in cyclotrons and other machines. A campaign to provide an initial supply of curium-244 for investigational purposes was concluded this year, after 10 kilograms of plutonium-239 had been exposed to neutron radiation for several years, resulting in the formation of about four hundred grams of curium-244. This material has been puri-

fied and will permit the determination of the macroscopic physical and chemical properties of curium on a much more accurate basis than has been possible in the past.

The shorter-lived isotope curium-242 is also of interest, primarily as a heat source. To produce it, a supply of americium-241 is required. This isotope, in turn, is formed by the beta decay of plutonium-241. Kilogram quantities of americium-241 have been made available for research or isotope production purposes. Although the production of curium-242 by neutron irradiation of americium-241 is a straight-forward procedure, the subsequent purification of the curium-242 and its fabrication into heat sources pose formidable problems. Most of the problems are the result of the intense heat and alpha radiation experienced as gram quantities of curium-242 are accumulated.

Requirements for fission product heat and radiation sources in the time period beyond 1968 may require a special plant (Hanford Isotopes Production Plant) for their manufacture. The projected capacity of such a plant and the estimated price of its products when operating at full capacity have been reported[12]. Although the magnitude of radioisotope production in the fission process or by neutron irradiation cannot be approached by machine irradiation, nonetheless, the production of radioisotopes by cyclotron bombardment has become increasingly important -- particularly for neutron-deficient isotopes. By the end of 1963, the United States was using 50% of the operating time of a 2.18 meter cyclotron[13] (originally built for research purposes) for the production of radioisotopes. Plans are being made for a more efficient and versatile accelerator for this purpose.

THERMAL APPLICATIONS OF ISOTOPES

One major thermal application of radioisotopes has been mentioned already; namely, the use of radioisotopes as heat sources in direct conversion electrical power supplies. More as a prediction of things to come than as a historical statement of accomplishments made, I would like to mention a few other thermal applications which appear promising and which are under study at this time in the United States. We have begun to examine the feasibility of employing radioisotopes as the source of energy in small rocket motors. Three concepts are under consideration: in the first, the heat evolved from the decay process is used to raise the temperature of a propellant gas in a direct convective heat transfer mode; in the second, the heat activates a catalyst which accelerates decomposition of hydrazine monopropellant; and in the third, radiation heating of the walls of the flow passages by means of a gamma-emitting isotope is used to raise the temperature of the propellant gas. Rocket motors of these types are characterized by their very high specific impulse but low thrust, and would, therefore, be useful in the upper stages of small, unmanned space vehicles destined for long trips to other planets or which are moving from a low altitude earth orbit to a high altitude orbit on a minimum energy trajectory.

Small radioisotope heat sources are being considered for use in highly instrumented satellites to keep the electronic components at an optimum, uniform operating temperature.

LARGE SOURCES FOR RADIATION PROCESSING

Another outlet for massive quantities of radioisotopes is expected to be found in the expanding radiation processing industry. It is now apparent that the research conducted during the past decade on the use of radiation for industrial processing is bearing fruit. This is evident with the appearance of several commercial radiation processes such as the production of ethyl bromide[14]; cross-linked polyethylene film, wire, tubing and related products[15]; medical supply sterilization[16]; and the production of semiconductor components[17]. Even more encouraging, however, is the greater number of radiation processes approaching commercialization. Included are the production of wood-plastic materials[18], curing of coatings, improved semi-conductor devices[19], textiles, "biodegradable" detergents[20], polymerization of ethylene and copolymers, and food processing[21].

In some cases, radiation produced by machines such as electron linear accelerators, resonant transformers, cathode ray tubes, and other devices prove to be superior from an economic viewpoint to the radiation from isotopes. Because of the early state of the radiation industry, it is not yet clear which source of energy will prove most acceptable for each application.

RADIATION TREATMENT OF FOODS

Treatment of selected foods with ionizing radiation for preservation or public health control measures is being seriously pursued in over 15 countries. National interests in this developing technology have grown rapidly in the past ten years. Research programs are, of course, concerned with items of promising economic value to each particular country. While radiation sterilization of food is important for special purposes involving long-term storage, the principal effort is on consumer products and is concentrated on the use of less than sterilization doses. Such effects include the inhibition of sprouting of potatoes and onions, the control of Salmonellae in egg or animal feed products, the inhibition of ripening of certain fruits, the disinfestation of grain and the extension of marketing or "shelf" life, through control of various spoilage microorganisms in food products such as meat, vegetables, fruits, fish and poultry[22,23,24].

Marine products are of particular international interest and research has been carried out with less than sterilizing doses of radiation on a wide variety of such products. The goal is to provide to the public a higher quality fresh fish or marine product, while also extending the marketing time. Lean fish, such as haddock and flounder, are typical subjects but interest also extends to clams, oysters, crab, shrimp and other marine products. Various approaches are used: (1) The treatment of fish at sea or at time of landing to maintain quality for distribution in the fresh state or prior to freezing; (2) Treatment to control such pathogens as the Salmonellaes, particularly in fish meal, or C. botulinum, type E, in smoked products; and (3) Treatment of frozen fish to maintain quality during thawing and subsequent distribution.

Radiation preservation of a variety of fruits has been investigated by a number of countries with increasing indications of several ultimately successful commercial processes. Strawberries have consistently shown promise. Although the marketing life of the berries is extended somewhat, the chief attainment is a marked decrease of loss of quality due to mold growth during distribution. Oranges, peaches, nectarines and sweet cherries show similar promise. Radiation treatment of fruits, such as papayas, mangoes and pineapples, are being investigated by countries with a semi-tropical climate. Not only are certain pests controlled through the interruption of their reproductive cycle, but plant pathogens can be destroyed. This would permit removal of quarantines and increased flow of trade for such products. Furthermore, radiation alters the physiology of the fruits, resulting in a decrease in rate of ripening and an increase in marketing life.

Human consumption of potatoes treated with low doses for sprout inhibition was approved in Canada in 1960 and, according to reports, earlier by Russia and Poland. In the United States, the use of certain types of radiation for disinfestation of wheat and wheat products and the sterilization of canned bacon has been approved for commercial use. Other countries such as France and the United Kingdom are developing protocols for certain irradiated foods, which hopefully will lead to clearances for commercial use in the future. Food irradiation technology is rapidly reaching the point where limited commercialization will take place. No extensive "breakthrough" is expected, but a steady growth will take place in this new food preservation method.

TRACER METHODOLOGY AND APPLICATIONS

The use of radioactive tracers in almost every phase of science and industry has become so commonplace that one can no longer hope to be all-inclusive in describing advances in the subject. Other speakers at this Conference are scheduled to discuss the use of tracers in such specific fields as agriculture, biology, medicine and hydrology. I will, therefore, limit my remarks to a few comments on new techniques which have rather general utility.

Tagging of a broad variety of materials, without regard to their chemical or physical form, may be accomplished by incorporating krypton-85 into the solid substance to be labeled[25,26]. This is performed by bombardment of the material with ionized krypton gas or by diffusion of the gas into the material under high temperature and pressure. The krypton gas at present contains 5% krypton-85; as the technique finds new applications, demands for higher concentration of the radioisotope will be made so that increased sensitivity of measurement will result.

The versatility of the use of krypton-85 as a tracer lies in the fact that almost any material may be labeled with it or "kryptonated." Already some 80 different solids, including elements, alloys and inorganic and organic compounds have been impregnated with the radioactive gas. However, the degree to which absorption of the gas occurs is a function of the physical structure of the material. Pyrolitic graphite, for example, may be labeled with a specific activity up to one curie per gram, whereas the absorption of gas in a steel sample will occur only within a few microns of the surface and the bulk specific activity then corresponds to microcuries per gram or less.

A substantial list of established applications of this technique exists, including the measurement of erosion of turbine blades, wear of bearings, and the kinetic properties of refractory metals at high temperatures[27]. If the krypton is incorporated into a reactive compound, then the extent of chemical reaction can be measured by determination of the amount of released radioactivity. This "radio-release" technique is similar to those reported previously for carbon-14 and hydrogen-3 compounds[28]. A method using krypton-85 is being developed for use in our space program as part of devices for measuring the oxygen content of the atmosphere of Mars, and for fluorine and hydrogen detectors for use in manned space vehicles and at launch sites.

At the last Geneva Conference, the first report was made of the total count method for determining the flow rate of fluids by use of radioactive tracers. Since that time, the method has found widespread use throughout the world and has become greatly diversified[29].

The advantages of using noble gas radioisotopes are demonstrated by another tracer technique reported recently[30]. Here the 5.27 day xenon-133 radioisotope was employed in the conventional dilution method to measure gas flow rates in an organic chemical plant. Other methods for determination of the flow rates had proven to be of no avail because of the corrosive nature of the gases and because they carried entrained particulate matter. The xenon tracer was chemically inert in this unusual environment, and no corrections for tracer deposition or fractionation were necessary. Flow rates ranging from 50 to 500,000 liters/minute were measured with a standard deviation of less than 1%. Counting of gas samples was performed in a large well-type plastic scintillator with excellent efficiency.

A hydrocarbon alkylation plant presented an unusually difficult problem in regard to measuring flow of sulfuric acid through various reactors and circulating loops[31]. By making rapid injections of radiogold into the system and then measuring tracer concentrations in acid samples or tracer flow through pipes, four quantities were easily measured: acid circulation rates, volume of acid in system, replacement rate of acid and entrainment of acid in the hydrocarbon stream.

I mention these accomplishments as only representative of how well radioisotope technology can be adapted to engineering studies in industry.

In science, tracer methodology has reached the point of prosaic acceptance. A survey of the abstracts of national meetings of research societies shows a large number of papers presented annually, which make use of radioactive isotopes, the greatest concentration being in the fields of medicine, biology, biological chemistry and physical chemistry.

Recoil chemistry or "hot atom" chemistry occupies such a significant role in tracer methodology that an entire symposium was devoted to the subject by the IAEA in 1960[32], while a survey article which appeared in 1962[33] on this subject states that six or seven hundred papers had been published in the field of recoil chemistry since its inception. The survey article itself cited 101 references in the literature, most of which appeared in 1959 and 1960.

When it is advantageous to incorporate a radioactive tracer into a functional group or groups of an organic compound, the use of tritium (hydrogen-3) has found widespread use[34,35]. Many workers have employed the self-labeling method of Wilzbach[36], but more recently, a new technique has become available which has substantially broader applications[37,38,39]. Hydrogen-tritium exchange is effected by contacting the organic material to be labeled with a tritiated phosphoric acid-boron trifluoride complex. A general

review of the method and some of its applications has appeared recently[40].

Again, there is space here to give only one example of the elegance of the tracer technique in basic chemistry. Collins and co-workers have been able to measure the secondary isotope effect in the reaction of acetophenone-beta-C^{14} with 2, 4-dinitrophenylhydrazine[41]. The ratio of specific rate constants of labeled and unlabeled species proved to be 1.0085 $\pm$ .0004. By using the same procedure with mixtures of deuterated acetophenone and acetophenone-beta-C^{14}, acetophenone-alpha-C^{14}, and other carbon-14 labeled ketones, the same group of investigators were able to measure with C^{14} the isotope effect of deuterium substitution for hydrogen in the ketone structures in the reactions which form phenylhydrazones[42]. For the case previously cited, the net effect of deuterating the methyl group in acetophenone was to increase the rate constant by a ratio of 1.111. In other words, a radioactive tracer was employed to determine the effect of a stable isotope substitution.

We should not lose sight of the fact that the radioisotopes which are found in nature are often of value as tracers or as chronographs. The classical work of Libby and co-workers in employing carbon-14 measurements for dating of archeological samples was considered important enough to be honored with the Nobel Prize in 1960. Other naturally occurring isotopes, both stable and radioactive, are being utilized to ever greater extent as the measurement techniques for them become more sophisticated. A major international symposium was held on this subject in 1962[43] and several articles or papers have discoursed on the general applicability of isotopic measurements or analysis in cosmochemistry, meteoritics and geochemistry[44,45,46]. Notwithstanding the innumerable applications for radioactive tracers which have been developed already for the benefit of science and industry, much remains to be done. In cases where there may be concern over the use of radioactive tracers, such as in large field experiments involving release to the biosphere or into consumer products, "activation" tracing will find expanded use. This involves the use of stable tracers followed by neutron activation analysis or charged particle activation analysis to measure the stable tracer concentrations. Although a beginning has been made in this technique[47], refinement of the analytical measurements remains the outstanding problem.

ISOTOPE MEASUREMENT AND CONTROL SYSTEMS

Past conferences have dwelt at length on the use of isotopes as sources of radiation in all manners of instruments such as thickness gages, density gages, composition gages, liquid level indicators, etc.[48]. Devices of this nature are now routinely used by industry in many thousands of installations. In some cases, competing instruments have been eliminated. Still, we can be certain that utilization of isotopic measurement and control devices in industry is still in its infancy. As one of my colleagues has written, "There seems no limit to the ingenuity of instrument inventors in using the penetrating, scattering, or selective absorption of radiations to test materials rapidly and feed back control information required by today's automated systems"[49].

Extension of a radiation scattering principle into a new realm is demonstrated by a device for measuring the density of air as a function of altitude to heights greater than 40,000 meters. This system[50] employs beta-ray forward scattering for sensing the density of air between a krypton-85 source and an anthracene scintillation crystal joined to a multiplier phototube. Background radiation count rate from an identical detector is subtracted electronically from the primary signal. Sensitivity of both circuits is increased by discriminating against pulses outside an optimum region of the Kr^{85} beta spectrum. A DC voltage directly proportional to net count rate is used to modulate a radio transmitter. This system has been incorporated into a balloon-borne package which also measures air temperature and pressure by other means. Sequential readings of density, temperature, and pressure are then transmitted to a ground receiving station. A recent calibration flight resulted in collection of useful data between 21,300 and 37,500 meters.

Oceanography is occupying an increasingly greater proportion of the attention of scientists in the United States today. Again, as expected, nuclear instruments are being developed for measurements in the remote

frontier deep below the surface of the ocean. One of the more complex of these is termed Deep Water Isotopic Current Analyzer, or DWICA. It consists of a circular planar array of 16 scintillation detectors surrounding a centrally located source of radioactive solution (typically I^{131}). Periodic injections of solution are swept by the ambient ocean currents to one or more detectors. A computer system analyzes time of arrival of activity at each detector, calculates velocity and direction of flow, and prints out the information in digital form[51]. DWICA is designed to operate at a maximum depth of 1800 meters.

The most exciting recent discovery that offers to expand this field immeasurably has been that of Dr. Rudolph L. Mössbauer. Although the Mössbauer effect is of outstanding importance in basic areas of physics, encompassing fundamentals of nuclear and molecular structure, it is also finding uses in a more applied sense. A review of applications of the Mössbauer effect alone would be extremely lengthy. I will hence limit my remarks to a few examples of applications which have appeared recently in the United States, observing once again that the discovery and experimental verification of the Mössbauer principle of recoilless emission and resonance absorption of gamma radiation and its extension to practical measurement systems would not be possible but for the ready availability of a wide variety of radioactive and stable isotopes which exhibit the Mössbauer effect.

The most publicized experiment of recent times occurred when Einstein's principle of equivalence was confirmed by Pound and Rebka[52]. They measured the difference in frequency of gamma quanta emitted at a height of about 20 meters above the surface of the earth and at the surface of the earth. The difference, as predicted by Einstein, should have been about one part in 10^{16} per meter of altitude, and this was detected quantitatively by measuring the frequency shift of the resonance absorption peak in Fe^{57}. In other words, the difference in "weight" of gamma quanta at an altitude of 20 meters and on the ground was measured! As Goldansky said in a recent review article[53] on the Mössbauer effect, "By comparison with this splendid piece of research, even the most elegant experiments are bound to suffer ..."

The greatest field of application so far has been the study of the physical and chemical environments of the target nuclei, whose energy levels are affected by the local magnetic fields, charge densities, and electric field gradients. In this sense, we speak of "Mössbauer spectroscopy." By far the favorite target has been Fe^{57}, with applications ranging from ferrimagnets[54] to hemoglobin[55]. The next is Sn^{119}, a favorite especially of the Russian workers.

The Mössbauer effect also provides a tool in the study of lattice dynamics and diffusion[56], since the intensity, line shape, and temperature shift are governed by the "stiffness" of the target environment. Applications in this area are expected to increase as difficulties in theory and technique are overcome.

Two groups have proposed the use of the Mössbauer effect to determine the small relative velocities at which one spacecraft will approach another in a docking or rendezvous maneuver, or the closure rate at which a space vehicle approaches a celestial body such as the moon[57,58]. Such velocities are anticipated to be of the order of millimeters per second in the final moments of approach (separation of a few meters). At such time, the use of radar is no longer possible, and yet accurate control of speed and alignment is essential to prevent damage from shock or from malfunction of the coupling mechanisms.

On the other hand, the use of a Mössbauer source and absorber is probably limited to ranges of 30 meters, but at this distance the relative velocity may be in excess of the capability of Mössbauer measurement. Therefore, another nuclear ranging system of a more conventional variety, which depends on the inverse square change in count rate with distance from a point gamma-ray source, has been proposed as the intermediate between the Mössbauer sensor and radar[58].

INSTRUMENTATION IMPROVEMENTS

A minor revolution has occurred in the field of radiation detection and analysis since the time of the last Geneva Conference because of at least two general scientific achievements: the development and commercial production of whole families of solid state electronic devices or components, and the design of very fast data acquisition equipment and computers.

Crystal scintillators have become more refined and have increased in size. Although thallium-activated sodium iodide remains the workhorse of the field, other inorganic materials such as cesium iodide and calcium iodide are receiving increased attention. A recent paper reported[59] that calcium iodide produces light outputs twice as great as those in NaI(Tl) when exposed to gamma radiation, and that pulse height resolutions of 5.2% and 9.9% (full width at half maximum) for Cs^{137} and Co^{57} gamma rays have been obtained with this material. However, problems of crystal fabrication remain to be resolved before a widely useful material will be available.

Plastic scintillators up to 40 cm in diameter are now sold in the United States. The University of Miami is employing scintillators of this size to measure trace concentrations of radioactive materials in sea water at depths up to 10,000 meters. In a system of two scintillators and corresponding electronic equipment, coincidence counting of cascaded gamma radiation from cobalt-60 enables one to detect this isotope at concentrations as low as 10^{-18} curie per liter in the presence of high background due to the naturally occurring potassium-40[60]. Care must be taken, however, to operate at depths sufficient to reduce the cosmic ray background to negligible levels.

Liquid scintillators have advanced correspondingly, although they, like the plastic varieties, remain inferior to the inorganic crystals in light production and pulse height resolution. However, they are superior for detecting very low concentrations of gamma emitters or for internal counting of low-energy beta emitters. Several applications for large liquid scintillators have appeared, including a system for detecting less than one microcurie quantities of Co^{60} moving at a rapid rate past the detector[61], a detector for measuring tracer levels of radioactivity in chemical process streams[62], and an apparatus for measuring neutron-proton coincidences produced by antineutrino interactions with hydrogen or deuterium in the scintillator fluid[63].

The multiplier phototubes employed in conjunction with the various varieties of scintillators are being improved constantly to decrease transit time, increase electron multiplication, and increase electrical stability and life. A tube as large as 60 cm in diameter has been produced[64] which, to my knowledge, represents the extreme in size.

Other tubes are now available with multiplication factors up to 10^8, permitting the detection of single photoelectrons. High-speed counting tubes have been reported recently to exhibit rise times of less than 0.5 nanosecond (approaching the minimum theoretical value of about 0.2 nanosecond), transit times of less than 4 nanoseconds, and dark currents of 10^{-9} ampere[65].

The most striking advance, however, has been in the improvement of semiconductor detectors for all types of electromagnetic or particulate radiation. It is now possible to say that semiconductor detectors have come of age, although much remains to be done in improving their stability at ambient temperatures and their efficiency for counting beta and gamma radiation. Silicon and germanium have been studied to the greatest extent. Silicon is superior to germanium in the sense that its noise level at room temperature is low enough to permit it to be used without cooling. Germanium normally requires cooling to liquid air temperatures to obtain the greatest sensitivity. At the same time, germanium is superior to silicon for detection and resolution of high energy gamma radiation because of its higher atomic number. Both materials can be drifted with lithium ions to increase their resistivity. Sensitive depths up to 1 cm have been achieved in silicon surface barrier detectors by this technique, leading to detectors capable of better pulse height resolution than scintillation detectors for gamma radiation of less than 2 Mev. Of course, the counting efficiency of these semiconductor detectors remains in the vicinity of 1%. We can expect that new semiconductor materials will be investigated within the next few years which will extend the range of gamma energy and increase detection efficiency. Cadmium telluride and gallium arsenide appear to be particularly attractive in this respect[66].

Perhaps equally exciting to the nuclear physicist and chemist has been the appearance of advanced electronic equipment to accompany the improved detectors. Transistorized pulse height analyzers of up to 400 channels are part of the equipment of most of the larger nuclear laboratories in the United States today,

and for more complex systems in which coincidences, anticoincidences, or time-delay relationships are recorded, it is possible to obtain instrumentation with up to 20,000 channels of recorded information, each channel having storage of up to 10^5 counts. The subject of multiparameter analysis of this type is receiving great attention, since the capacity to record data has now exceeded the capacity to analyze the significance of the data. At a conference held in 1962 in the United States, applications of multi-parameter analysis in nuclear physics was discussed, and evidence of the progress being made in this complex field was substantial[67].

To the person interested in radioisotopes technology, the techniques of greatest interest are those in which a mixture of gamma spectra from the simultaneous decay of several isotopes is resolved into the constituent spectra, permitting quantitative identification of the isotopic mixture, or resolution of the complex decay mode of a radioisotope. Such procedures are of utmost importance in neutron activation analysis, and a number of laboratories are vigorously attacking the problem[68]. The approach being employed is first to assemble a "catalog" of the spectra of all isotopes of interest in a standard detector geometry and to record the information on punched or magnetic tape by means of an analogue-to-digital converter. An unknown spectrum is then recorded and stored in similar fashion, and a high speed computer is then employed to make comparisons of peaks, sort out individual spectra, and finally calculate the relative amounts of identified isotopes present. The time required to perform the analysis is thus reduced by several orders of magnitude in comparison to hand computations.

Excellent summaries of this and other applications of multi-parameter and computer analysis to radioisotope work have been published by several of the prominent workers in the field[69,70,71]. In general, the combination of pulse height analyzers and computer memories, plus fast and graphic readout systems, facilitates the performance of ordinarily laborious tasks such as calculation of the pulse-height response of a scintillation crystal as a function of gamma-ray energy, computation of coincidence sum spectra for coincident or cascade gamma emission, resolution of superimposed photopeaks, and angular correlations of coincident events.

One particular area that has benefited greatly from such advances in automation and instrumentation, has been nuclear activation analysis. In recent years, this important technique has grown from an infrequently used laboratory procedure to a method reknowned for its sensitivity and diversity of application[72,73].

THE FUTURE OF RADIOISOTOPE APPLICATIONS

Notwithstanding all of the advances made in the short period of six years, our optimism for the future is tempered with the sobering thought that much remains to be done before the potentials for the use of radioisotopes in science and industry are more fully realized. More detailed education of scientists and engineers, who are not nuclear specialists, is required to acquaint them with the unique characteristics and versatile capabilities of radioisotopes, as well as their already proven applications. Scientists and engineers so educated could employ radioisotope technology as a ready tool in the same manner as the more conventional techniques of chemistry, physics, and other basic disciplines.

Regulation of the use of radioisotopes by governmental agencies is still too restrictive in some instances. However, many gains have been made in extending the freedom of use of these materials while maintaining good safety practices. Industrial management has not yet fully recognized that the use of radioisotopes can provide substantial savings in time and money not possible by other means. This requires a continuing program of education at all levels.

Several predictions of technical advances to be realized have been made earlier. We can expect to see many megacurie quantities of specific radioisotopes in use for radiation processing, for small electric power generators and for space heat applications. We will see them become employed in more routine ways as well as in explorations from the bottom of the sea to outer space -- at the extremes of our environment. It has been said before, and it may need to be repeated for many decades, that the applications of isotopes will be limited only by man's ingenuity.

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